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Pollutant Sampler for Measurements of Atmospheric Acidic Dry Deposition

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16. ABSTRACT

An acidic pollutant sampler for dry deposition monitoring has been designed and evaluated in laboratory and field studies. The system, which is modular and simple to operate, samples gaseous HNO_3, NH_3, SO_2 and NO_2 and particulate SO_4^{-2}, NO_3^- and NH_4^+ and is made of Teflon to minimize trace reactive gas sorption. Particles greater than about 2μm are removed with a cyclone. A transition-flow reactor (TFR) Teflon tube contains a Nylon liner for collection of a constant fraction of gaseous HNO_3 and a Nafion liner for collection of a constant fraction of NH_3. A Teflon filter that removes the fine particle, a Nylon filter that collects the HNO_3 that penetrates the TFR and any from the decomposition of NH_4NO_3 and an oxalic acid coated glass fiber filter that collects the NH_3 that penetrates the TFR and any from the decomposition NH_4NO_3 are contained in a three-filter holder down stream of the TFR. The final section of the system contains two glass fiber filters impregnated with triethanolamine for SO_2 and NO_2 collection. Excellent results were obtained in week-long runs with parallel sampling. In the first seven of these week-long runs the average difference between samples for gaseous HNO_3 was 4.6% with a standard deviation of only 3.7. In the same sampling runs, the average difference for particulate sulfate was 3.0% with a standard deviation of only 1.4.

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Pollutant Sampler for Measurements of Atmospheric Acidic Dry Deposition

EPA/600/J-86/382
JOURNAL ARTICLE

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■ An acidic pollutant sampler for dry deposition monitoring has been designed and evaluated in laboratory and field studies. The system, which is modular and simple to operate, samples gaseous HNO_3, NH_3, SO_2, and NO_2 and particulate SO_4^{2-}, NO_4^-, and NH_4^+ and is made of Teflon to minimize trace reactive gas sorption. Particles greater than about 2 μm are removed with a cyclone, which is followed in the system by a transition flow reactor (TFR) containing a nylon liner for collection of a constant fraction of HNO_3 and a Nafion liner for collection of a constant fraction of NH_3. The TFR is followed by a three-filter holder containing, in order, a Teflon filter to collect the fine particles, a nylon filter to collect HNO_3, and an oxalic acid impregnated glass-fiber filter to collect NH_3. The backup nylon and oxalic acid filters collect the gaseous HNO_3 and NH_3 that penetrated the TFR and that from the decomposition of the NH_4NO_3 collected on the Teflon filter. The final section of the system contains two glass-fiber filters impregnated with triethanolamine for SO_2 and NO_2 collection. The analyses for HNO_3, NO_3^-, NO_2, SO_4^{2-}, and SO_2 are done by extracting the exposed collectors and running aliquots on an ion chromatograph. The NH_3 and NH_4^+ are determined by either a specific ion electrode or the indophenol autoanalyzer colorimetric method. Results from both laboratory evaluation and field studies are presented. In 7-week-long studies, the average difference between samples from parallel runs for gaseous HNO_3 was 4.6% with a standard deviation of 3.7.

Introduction

The difficult problems in the measurement of the dry fluxes of atmospheric acids and their precursors preclude long-term routine direct monitoring of dry deposition. A practical alternative method is to estimate dry deposition by the "concentration monitoring" method described by Hicks et al. (*1*). In this method, the dry flux, F, is estimated from $F = v_d\bar{C}$, where v_d is the average deposition velocity estimated for the sample-averaging period used to determine the average concentration $\bar{C}$ (*2*). Protocols for estimating v_d and sampling to determine C for estimating weekly average deposition rates for the National Trends Network are being developed (*3*). For protocol development and monitoring, pollutant cumulative samplers are required that can operate for periods of about 0.25–7 days.

In some sampling systems, the concentrations of important gases such as HNO_3, NH_3, O_3, and H_2O_2 may be reduced by passing them over particles collected on a filter. Also, the evaporation of NH_4NO_3 may increase the air concentrations of HNO_3 and NH_3 after the filter. Our approach is to eliminate these biases by removing a constant fraction of these reactive gases in a "transition flow reactor" (TFR) in front of the filter (*2*). In this paper, we report laboratory and field performance sampling results for a combination of the TFR, filters, and coated filters to sample free HNO_3 and NH_3, particulate SO_4^{2-}, NO_3^{2-}, and NH_4^+ ions, penetrating and evaporating HNO_3 and NH_3, and SO_2 and NO_2. We have not yet developed the sections for sampling H_2O_2, O_3, and large particles.

Experimental Section

Sampling System. The samping system (see Figure 1) is made of modular units arranged in series to collect the various pollutants of interest in acidic dry deposition monitoring. Before the detailed descriptions of each sampling module, a brief discussion of the sampling design rationale follows. To minimize possible trace reactive gas sorption, the entire sampling system except collection surfaces is made of Teflon. The first unit is a cyclone for removing the particles larger than about 2-μm aerodynamic diameter. This is followed by a vortex remover to eliminate the cyclonic flow setup in the cyclone and by a transition-flow reactor (TFR) (*2*, *5*) tube that contains a nylon strip for the collection of a constant fraction of the gaseous HNO_3 and a Nafion strip (*5*) for collection of a constant fraction of NH_3. Nafion is a modified Teflon that has sulfonic acid groups added through side chains for ion exchange. The TFR is followed by a three-filter holder that contains in order a Teflon filter to collect the fine particles, a nylon filter to collect the remaining HNO_3 and an oxalic acid impregnated glass fiber filter to collect the remaining NH_3. This is followed by a flow splitter. The low flow side contains a series of two glass-fiber filters impregnated with triethanolamine (TEA) for SO_2 and NO_2 collection. Following the TEA filters is a 1.78 standard L/min mass flow controller. The higher flow side contains only a 14.3 standard L/min mass flow controller. The final component is the pump. Figure 1 is a schematic diagram of the entire system.

Cyclone and Vortex Remover Module. An all-Teflon cyclone based on the Southern Research Institute design for cyclone II described by Smith et al. (*6*) is used to remove the coarse particles. When both sides of the sampling system are operating, the flow through the cyclone is 33.2 standard L/min. With this flow, the cyclone has a D_{50} of about 1.8 μm. With only one side sampling, the flow is 17.3 standard L/min and the D_{50} is about 2.5 μm. The cyclone is mounted so that the entrance is horizontal; a special removable entrance cap protects the system from the intake of rain. A nomimal flow rate of 1 L/min out the bottom of the stagnation zone of the cyclone through a filter and critical orifice removes the excess large particles and prevents their accumulation in the event of a heavy particulate episode caused by high winds or dust storms. On the top of the cyclone at the exit is a vortex remover to straighten the flow before the gas stream enters the TFR. A Teflon tee was found to be adequate for this purpose. However, the 90° change in flow sets up an oscillating flow that must be eliminated. A straight flow section of eight tube diameters was found to give the desired plug flow. Above eight tube diameters, no difference in collection efficiency was found, and since a minimum length in the overall system is desirable for shipping and installation, the shortest possible length was used. To determine if the cyclone and vortex remover interfered with the collection of gaseous HNO_3, runs were made on the collection of HNO_3 with and without the cyclone/vortex remover in the system. No difference was seen for the collection efficiency of HNO_3. No particles were present in these runs.

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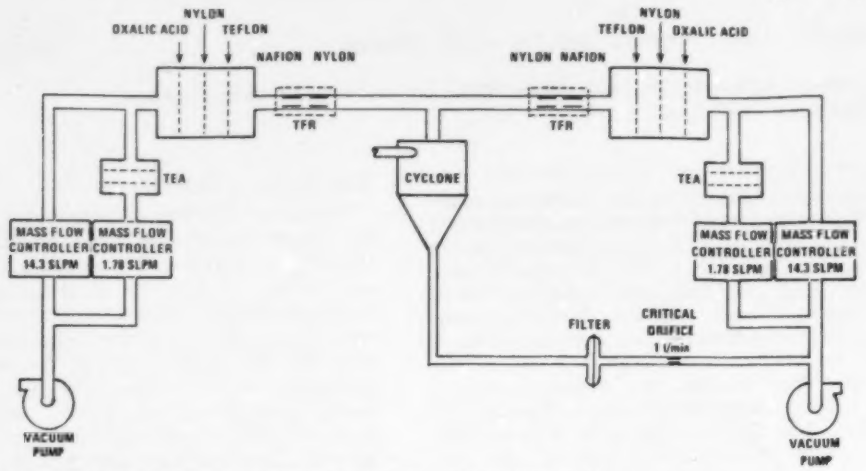

Figure 1. Schematic diagram of concentration monitor for estimating acidic dry deposition.

TFR Module. The TFR is a 0.953 cm inside diameter Teflon tube that holds the 3.2 cm long nylon and Nafion wall liners. The nylon liner's leading edge is located 8 cm from the vortex remover and is positioned on the inner circumference of the tube. The Nafion linear is positioned on the downstream side of the nylon linear with a small-ridged stop separating the two strips. The flow in the TFR is maintained at 16.1 standard L/min with mass flow controllers. An all-Teflon valve will be added to isolate the TFR and the three-filter holder when the two sides of the concentration monitor (CM) are run at different times such as day–night sampling. This isolation will prevent back-diffusion of HNO_3 from possible NH_4NO_3 and other nitrate decomposition when that side is not sampling.

Three-Filter Holder and TEA Filters Modules. The all-Teflon holder contains the Teflon filter for fine particle removal, the backup nylon filter to remove all the gaseous HNO_3 that penetrated the TFR and any that is formed from the decomposition of NH_4NO_3 and other nitrates, and the oxalic acid impregnated glass-fiber filter to remove all the NH_3 that penetrated the TFR and any that formed from the decomposition of the NH_4NO_3. The oxalic acid filters are prepared by immersing the glass-fiber filters in a solution of 5% oxalic acid in methanol. The immersed filters are allowed to drain for a few seconds, and then the excess solution is removed by aspirating for about 1 min in a fritted Buchner funnel. Two TEA filters are mounted in separate Teflon filter holders. Better results were obtained in our laboratory evaluation with this arrangement than having two filters in one holder. The TEA filters are prepared by saturating cleaned glass-fiber filters with a solution of 25 g of TEA and 4 g of ethylene glycol in 25 mL of acetone and diluted to 100 mL with deionized water. The excess solution is removed as described above for the oxalic acid filters. Before treatment, the filters are cleaned in hot (80 °C) 5% KOH solution and rinsed several times with deionized water. The excess water is removed and the filters are immediately treated with the TEA solution.

Flow System. In transition flow collection, the mass flow must be held constant to maintain a constant collection efficiency. Mass flow controllers were used in all the tests. The systems used were 30 L/min controllers (Tylan model 262) for the 14.3 standard L/min legs and 5 L/min controllers (Tylon model 260) for the 1.78 standard L/min legs.

Analytical Procedures. The six types of active surfaces in the CM were analyzed as shown in Table I. The Teflon filter was halved before extraction because of the differing extraction solution needed for anion and cation analyses. Extraction for ion chromatographic (IC) analysis was performed by adding 10–20 mL of IC anion eluant to a sample vial containing the filter or strip and sonicating or vortexing for 15 min. Extraction for NH_4^+ analysis by ion-selective electrode was performed by adding 10–20 mL of 0.1 N NaOH to the sample vial containing the filter or strip and sonicating for 2 h. For best quantitation the mass of each ion collected by the sampler should be at least 10 times larger than the detection limit shown in Table I.

Table I. Analytical Requirements

	SO_4^{2-}	NO_3^-	NO_2^-	NH_4^+
nylon strip (3.2 × 3.8 cm)	X	X		
Nafion strip (3.2 × 3.8 cm)				X
Teflon filter (47-mm diameter)	X	X		X
nylon filter (47-mm diameter)	X	X		
oxalic acid filter (47-mm diameter)				X
TEA filters (47-mm diameter)	X	X	X	
method	IC[a]	IC	IC	ISE[b]
detection limit, μg[c]	0.25	0.6	0.5	0.6

[a] Ion chromatography. [b] Ion-selective electrode (The indophenol method is also acceptable and has a detection limit of about 0.9 μg). [c] Based on a 10-mL extraction.

Discussion and Results

Transition Flow Reactor (TFR) Evaluation. For a TFR that collects a constant fraction F of the reactive gas species entering, the ambient concentration, $C(0)$, can be determined from the measurements of the collected mass, m, and the sample volume, V, by

$$C(0) = m/(VF) \quad (1)$$

The theory and preliminary performance of the TFR for collecting gaseous HNO_3 have been described by Durham and Ellestad (2). The TFR's collection efficiency for a reactive gas such as HNO_3 being deposited in a tube operated in transition flow ($2100 < Re < 3500$) is

$$F = 1 - [C(x)/C(0)] = 1 - \exp[-2\pi rDx/(\lambda Q)] \quad (2)$$

where F = fraction collected, Re = tube Reynolds number, $C(0)$ = the reactive gas concentration at the inlet (μg/m^3), $C(x)$ = the reactive gas average concentration at the axial distance x (μg/m^3), r = the radius of the tube (cm), D = the gaseous diffusion coefficient of the reactive species (cm^2/s), x = the axial distance from the inlet of the reactive zone (cm), λ = the fictitious stagnant air film thickness (cm) and Q = volumetric flowrate (cm^3/s).

Table II. Collection Efficiency (F) of Gaseous Nitric Acid in TFR

flowrate, std L/min	F, fraction collected	D, cm^2/s	λ, μm	no. of observations
13.4[a]	0.100 ± .004	2.76 ± 0.11	435 ± 20	12
16.1[a]	0.089 ± 0.004	2.93 ± 0.14	410 ± 20	10
16.1[b]	0.088 ± 0.016			6
17.8[a]	0.087 ± 0.008	3.18 ± 0.31	375 ± 40	12

[a] From Durham and Ellestad (2), 2% dry test meter correction factor applied. [b] Results obtained by Battelle Laboratories, Columbus, OH.

Table III. [HNO_3] Measured at Oak Ridge by Batelle in June 1984

run	sampling time, h	side A, μg/m^3	side B, μg/m^3	av, μg/m^3	% diff from av
1	24	2.1	1.1	1.6	31
2	31	1.4	1.7	1.6	9.7
3	47	1.3	1.5	1.4	7
4	48	1.7	1.2	1.4	17
					av 16[a]

[a] σ = 9.3.

The fictitious film does not have a sharp boundary and λ cannot be calculated directly from theory. Since λ decreases as Q increases, the TFR must be experimentally calibrated for practical applications.

Changes in temperature (T) and in atmospheric pressure (P) change the diffusion coefficient and the volumetric flowrate, which cause the collection efficiency to change. However, control of the mass flowrate removes from the ratio (D/Q) in the exponent in eq 2 the influcence of pressure change and reduces the influence of temperature change to $(T/298)^{0.5}$, where the reference temperature for D is T = 298 K. For a temperature range of −20 to +40 °C, the variation in the TFR's collection efficiency is $\Delta F/F$ = 0.06, where F is the midpoint of the range. F may vary during sampler operation if the reactive gas forms molecular complexes such as hydrates. The possible formation of $HNO_3 \cdot nH_2O$ complexes, in which n increases as relative humidity increases was considered as a possible bias to D. However, the diffusion coefficient of HNO_3 in moist air has been shown to be constant (0.118 ± 0.003 cm^2/s), and thus, n is constant, over the relative humidity range of 5 to 95% (*4*). The measured value of D corresponds most closely to that expected for n = 0, which is nonhydrated HNO_3. Thus, pressure and relative humidity (5–95%) variations do not bias F, and expected variations in temperature (less than ±30 °C) will impose less error on F than the chemical analyses.

We report here the results of laboratory and field testing of the system to evaluate the variability of the TFR's efficiency F and the collection by the filter sections. Laboratory performance of the TFR for HNO_3 sampling was investigated by Battelle Laboratories (Columbus, OH) and by us. We observed that the dependence of F on the mass flowrate Q_m was the least for Q_m of 16.1 standard L/min, which corresponds to Re ~ 2500. The results of the measurements of F as a function of flowrate, presented in Table II, demonstrate that the TFR's collection efficiency is stable at constant mass flowrate in the range 13.4–17.8 standard L/min and is reproducible between laboratories. The change in F is weakly dependent on change in Q_m. For the range of Q_m given in Table II, $(\Delta F/F)/(\Delta Q_m/\bar{Q}_m)$ is about 0.5, where ΔF and ΔQ_m are the range intervals and F and Q_m are the range midpoints. The fluctuations of the mass flowrate Q_m are controlled within ±2%, which transmits less than 1% variation to F.

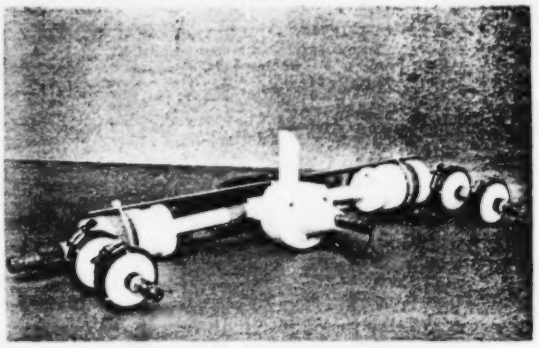

Figure 2. Concentration monitor.

Table IV. [HNO_3] Measured at RTP, NC, August–October 1984

run[a]	μg/m^3 side A	μg/m^3 side B	μg/m^3 av	% diff from av
1	2.43	2.94	2.68	9.5
2	1.41	1.26	1.34	5.6
3	1.24	1.36	1.30	4.6
4	2.65	2.63	2.64	0.2
5	2.13	1.75	1.94	9.8
6	3.30	3.20	3.25	1.6
7	1.90	1.87	1.88	0.8
				av 4.6[b]

[a] All runs were for 7 days. [b] σ = 3.7.

Table V. [SO_2] Measured at RTP, NC, August–October 1984

run	μg/m^3 side A	μg/m^3 side B	μg/m^3 av	% diff from av
1	4.43	3.93	4.18	6
2	1.27	1.21	1.24	2.4
3	3.81	3.03	3.42	11.4
4	4.96	5.23	5.10	2.5
5	2.26	2.30	2.28	0.9
6	8.44	8.18	8.31	1.6
7	1.91	2.54	2.22	14
				av 5.6[a]

[a] σ = 4.85.

Total System Field Evaluation. Battelle ran a modified system including only the cylone, vortex remover, TFR (for HNO_3 only), and fine particle filter to evaluate the system's variability. The results for the HNO_3 collection by the TFR in this study are given in Table III. These data are for a dual system with the sides designated A and B. The percent differences for the four runs was 16% with a standard deviation of 9.3.

The prototype shown in Figure 2 has been run for several continuous week-long tests. The results of the first 7-week-long tests are given in Tables IV–VII. Table VIII gives particulate NH_4^+ data for the first four tests. The samples for the other tests were not analyzed for NH_4^+. Good agreement was obtained between the two sides designated as A and B in the tables for all pollutants measured. The greatest differences were seen with the SO_2 and NO_2 system when the concentrations were below 5 μg/m^3. The average difference for gaseous HNO_3 was 4.6% with a standard deviation of only 3.7. Similarly, the particulate sulfate had excellent agreement with an average difference of 3.0% and a standard deviation of only 1.4.

Table VI. [NO_3^-] Measured at RTP, NC, August–October 1984

	μg/m³			
	side			
run	A	B	av	% diff from av
1	7.88	7.77	7.82	0.7
2	5.88	7.14	6.51	9.7
3	4.71	3.77	4.24	11
4	6.14	6.88	6.51	5.7
5	3.22	4.97	4.10	21
6	15.8	17.9	16.8	6.3
7	7.20	8.02	7.61	5.4
				av 8.6[a]

[a] $\sigma = 6.0$.

Table VII. Particulate SO_4^{2-} Measured at RTP, NC, August–October 1984

	μg/m³			
	side			
run	A	B	av	% diff from av
1	10.4	9.56	10.0	4.4
2	2.00	2.11	2.05	2.7
3	6.48	6.34	6.41	1.1
4	3.74	3.93	3.84	2.5
5	3.24	3.45	3.34	3.2
6	8.71	9.01	8.86	1.7
7	6.08	5.44	5.76	5.5
				av 3.0[a]

[a] $\sigma = 1.41$.

Table VIII. Particulate NH_4^+ at RTP, NC, August–September 1984

	μg/m³			
	side			
run	A	B	av	% diff from av
1	2.80	2.93	2.86	2.3
2	0.71	0.68	0.70	2.2
3	1.48	1.47	1.48	0.4
4	2.77	2.55	2.66	4.2
				av 2.3[a]

[a] $\sigma = 1.34$.

HNO_3 blanks on the nylon filters and strips were investigated. The blank values obtained for runs 3–5 are given in Table IX along with the concentrations from these runs. As can be seen from these data, the blank HNO_3 accounts for only 0.25% of the amounts collected; thus, there is no blank problem.

Some batches of nylon filters collect small amounts of SO_2, typically about 5 μg per 47-mm filter. If this amount was large, it could interfere with the SO_2 collection. Both in the tests at EPA and those reported by other investigators (*7*, *8*) the amount of SO_2 per 47-mm filter appears to reach a maximum of about 5 μg. When this is compared to an average collection in the 47-mm filter of 250–500 μg of SO_2, only a 2% or less error results. The results of several tests at EPA are shown in Table X. These results and those reported by Hansen (*7*) suggest that there is a limited capacity of about 5 μg of SO_2 on a 47-mm diameter nylon filter. In the field studies we have seen more than 5 μg as SO_4^{2-} collected on both the nylon strips and filters. These results are in agreement with Eatough who has shown that nylon collects dimethyl and monomethyl sulfates. He is now developing a sampling system for these compounds with a nylon collector. Batches of nylon filters

Table IX. HNO_3 Blanks

	μg/filter			μg/filter		
	side			side		
run	A	B	av	A	B	av
3	0.81	0.61	0.71	185	185	185
4	0.42	0.28	0.35	204	192	198
5	0.35	0.50	0.42	183	238	210
			0.5			198

0.5/198 × 100 = 0.25%

	μg/strips			μg/strip		
	side			side		
	A	B	av	A	B	av
3	0.1	0.1	0.1	22.5	20.4	21.4
4	ND	0.1	0.05	39.0	39.2	39.1
5				25.3	30.7	28.0
			0.075			29.5

0.075/29.5 × 100 = 0.25%

Table X. SO_2 Collection by Nylon, Teflon, and Carbonate

	μg (as SO_4^{2-})		
run	nylon	Teflon	carbonate
4	1.7	0	19.4
6	1.8	0	a
7	4.5	0.7	424
10	1.2	0	18.6
12	3.9	0.2	446

[a] Sample lost; exposure time the same as run 4.

received by other investigators (*9*) after the completion of this study have shown much higher SO_2 collection. The use of such filters would cause significant errors in the SO_2 measurements.

The limit of detection based on an assumption of 0.05 μg/ml sensitivity for NO_3^- by IC is 0.03 μg/m³ for a 7-day exposure. The values are 0.8 μg/m³ for 6 h, 0.4 μg/m³ for 12 h, and 0.2 μg/m³ for 24 h. With careful extraction and analysis, sensitivity of 0.005 μg/mL has been obtained in our laboratory. This value will yield a lower detection limit of 0.5 μg/m³ for a 1-h sample. The capacity of the system is good and can handle a 7-day sample with an average concentration of over 15 μg/m³.

Conclusion

A workable dual sampling system for measuring the concentration of acidic dry deposition pollutants for sampling periods of 1 h to 7 days has been developed and field tested. The system can measure gaseous HNO_3, SO_2, NO_2, and NH_3 and particulate SO_4^{2-}, NO_3^-, and NH_4^+. It is modular, simple to operate, easy to install and ship, and the collectors can be easily analyzed by either existing IC or automatic colorimetric methods.

Acknowledgments

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Registry No. TEA, 102-71-6; NH_3, 7664-41-7; SO_2, 7446-09-5; HNO_3, 7697-37-2; NO_2, 10102-44-0; SO_4^{2-}, 14808-79-8; NO_3^-, 14797-55-8; NH_4^+, 14798-03-9; Teflon, 9002-84-0; oxalic acid, 144-62-7; Nafion, 39464-59-0.

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